

# Magnetostriction of Polycrystalline Strong-Textured Fe-17 at% Ga Laminates

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Magnetostrictive bulk Fe-17 at%Ga alloy was fabricated by combining laminates of rapid-solidified ribbons (80  $\mu\text{m}$  in thickness) using the spark plasma sintering/joining (SPSJ). The SPSJ is characterized by a short time, low temperature heating and sintering process. The laminated sample made by the SPSJ maintained a unique metallurgical microstructure of polycrystalline texture of columnar grains, as well as the almost non-equilibrium metastable phase with little evidence of the precipitates of the ordered phases as found in the as-spun ribbons. An excellent sintered sample exhibiting large magnetostriction was obtained under a condition of compressive stress of 100 MPa at a temperature of 973 K. The magnetostriction depended on compressive pre-stress level for each specimen and reached about 100 ppm, which was half the value obtained for the ribbon sample. Furthermore, by subjecting this specimen to a short annealing process, the magnetostriction increased to 170 ppm, comparable to the value for the ribbon.

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## 1. Introduction

Magnetic, magnetostrictive and elastic properties of  $\text{Fe}_{100-x}\text{Ga}_x$  ( $13 \leq x \leq 35$ ) single crystals were studied by A. E. Clark *et al.*, and they found that the  $\text{Fe}_{100-x}\text{Ga}_x$  ( $17 \leq x \leq 19$ ) show good durability, high ductility and large magnetostriction of 400 ppm ( $\times 10^{-6}$ ) in a low magnetic field.<sup>1-3</sup> However, saturation magnetostriction of this alloy system decreased at higher concentrations of gallium ( $19 < x$ ). In the equilibrium phase diagram of Fe-Ga alloy system, where the phase transformation from the bcc disordered A2 phase to the fcc ordered  $\text{L}_{12}$ , as well as to the bcc ordered  $\text{D}_{03}$  phase appears at the higher concentration of gallium. The precipitates of the ordered phases generally suppress the microscopic magnetic domain movements and thus influence the magnetic and magnetostrictive properties of the disordered Fe-Ga alloys. If the disordered phase at the high-temperature region can be frozen at room temperature without precipitating the ordered phases, larger magnetostriction can be expected. As is known, the advantages of using the melt-spinning method are extension of solid solubility, grain refinement, reduction or elimination of microsegregation, and formation of the metastable phase. These bring about improvements in strength toughness, hardness, wear resistance, heat resistance and corrosion resistance. Attention is especially focused in this study on the formation of a non-equilibrium condition that might suppress the ordered phases. Moreover, microstructures from dendritic to amorphous can be manufactured by changing the cooling rate, and anisotropic columnar grains are formed at a medium cooling rate.<sup>4</sup>

The basic approach begins by developing a high performance magnetostrictive actuator/sensor Fe-Ga alloy using the rapid-solidification process, which is schematically shown in Fig. 1. This is the key process of the first stage that allows a unique non-equilibrium metastable microstructure to be

obtained by the rapid-solidification, in comparison to normal equilibrium phase melt-worked and annealed single and polycrystalline bulk samples (see upper-right micrograph in Fig. 1<sup>5</sup>).

Based on the above-mentioned situation, the melt-spinning method was applied to develop rapidly solidified thin Fe-Ga ribbons. These ribbons had fine columnar grains with a strong [001] oriented texture, and maximum magnetostriction when a magnetic field was applied to the direction nearly normal to the plane (Fig. 2(b),  $\theta = 80^\circ$ ), that being the direction of easy magnetization of the texture along the thickness direction.<sup>6</sup> The term 'maximum magnetostriction' means the observed maximum strain at the maximum applied magnetic field. However, since the mass of the ribbon sample was very small, no large driving force would be available from the actuator for engineering purposes. In order to overcome this practical problem and extend the applicability of the high performance ribbon sample, the spark plasma sintering/joining (SPSJ) method was applied to laminate the ribbons for fabricating more robust and bulk actuator/sensor materials.

In this study, we propose a novel and systematic material processing method to develop robust bulk sensor/actuator materials that could maintain the unique microstructure to provide rapid-solidified material elements with high performance material properties.

Figure 3(a) shows a schematic diagram of the SPSJ method. Punches are inserted into a fixed die which is filled up with samples. The samples are then set in a chamber and two electrodes are then inserted into them. Between these electrodes, spark plasma with high energy is created by pulsed direct current, and therefore the SPSJ is characterized by very short time and low temperature heat processing.<sup>7</sup> These features provide a distinct advantage in the bulk processing of rapid-solidified ribbons with the unique [001] oriented texture, as the SPSJ will work to suppress the precipitation of any ordered phase, as well as recrystallization of fine grains in the as-spun ribbon.

Magnetic and magnetostrictive properties of the Fe-Ga sintered samples that were fabricated by various compressive

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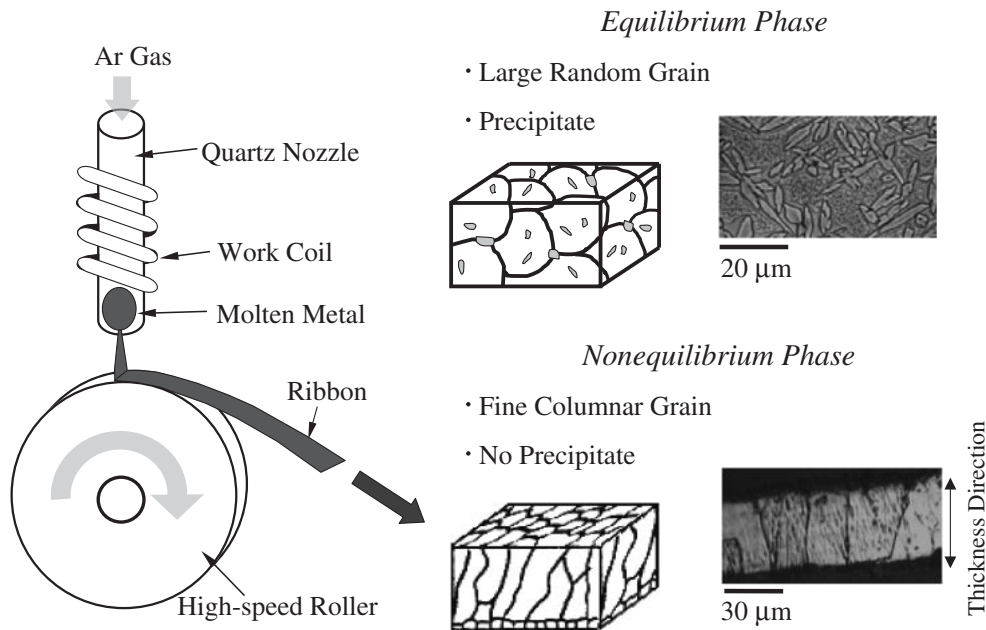


Fig. 1 Schematic diagram of the melt-spinning method and change of metallurgical microstructure. Upper and lower micrographs show the equilibrium phase with the precipitates<sup>5)</sup> and the unique non-equilibrium structure of Fe–Ga alloy, respectively.

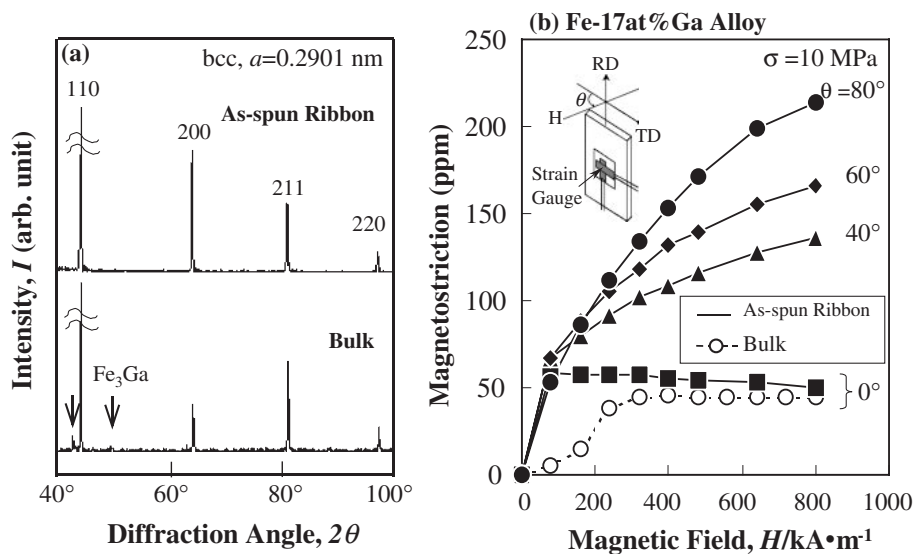


Fig. 2 (a) X-ray diffraction patterns and (b) dependence of magnetostriction on direction of Fe–17 at%Ga alloys. Arrows in the left figure show peaks of the ordered  $\text{Fe}_3\text{Ga}$  precipitates in the melt-worked bulk material.

stresses and sintering temperatures under SPSJ process conditions were investigated. We conclude that the developed bulk Fe–17 at%Ga laminates have a potential use as sensor/actuator materials which can nearly match the performance properties exhibited in as-spun ribbon elements.

## 2. Experimental Procedures

Ingots of Fe–17 at%Ga were prepared using an arc melting method by electrolytic iron (purity 99.999%) and gallium (purity 99.9999%). The iron and gallium were homogenized by holding the temperature at 1173 K for 24 h. Ribbon samples 80  $\mu\text{m}$  thick and 5 mm wide were produced using the single-roll melt-spinning method in an argon atmosphere

from a starting ingot. 200–300 sheets of the rapid-solidified ribbon were stacked and set in a tungsten-carbide die. The laminated ribbons were sintered under three different SPSJ conditions *i.e.* pressure of 50 MPa at 973 K, 100 MPa at 973 K and 300 MPa at 873 K. During the sintering process, each ribbon was held for 5 min at the sintering temperature. The temperature was raised at the rate of 20–25 K/min. The sintered bulk samples are approximately 40 mm long, 5 mm wide and 7–9 mm thick, where the thickness is in the direction normal to the ribbon plate.

X-ray diffraction (XRD) profiles of the surface for each sample were obtained using  $\text{CuK}\alpha_1$  radiation. Magnetization was measured using a vibrating sample magnetometer (VSM). Longitudinal magnetostriction parallel to the thick-

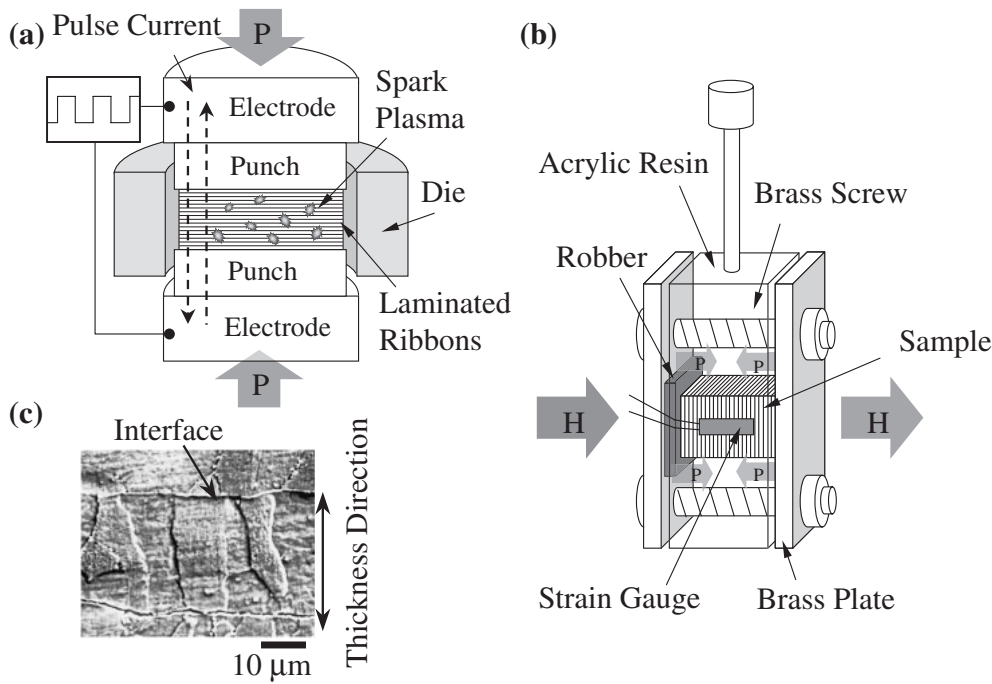


Fig. 3 Schematic diagram of (a) laminated ribbons during SPSJ, (b) magnetostriction measurement method by strain gauges for sintered sample, and (c) SEM micrograph of cross section of sintered sample (100 MPa, 973 K).

ness direction was measured using strain gauges. These measurements were done under the compressive pre-stresses of  $-20$ ,  $-60$  and  $-100$  MPa, using the apparatus shown in Fig. 3(b), where the compressive pre-stresses,  $\sigma$ , were applied in the direction parallel to the magnetic field. A polymeric rubber spacer plate whose stiffness was very small was inserted between the specimen surface and the brass plate holder to prevent excessive rigid restraint during measurement of magnetostriction. The value of  $\sigma$  was determined by multiplication of the pre-strain value by Young's modulus of elasticity for each sintered sample. Magnetostriction was determined by averaging the values obtained from the strain gauges on both surfaces.

### 3. Results and Discussion

#### 3.1 Structure and texture

##### 3.1.1 The effect of sintering pressure and temperature

X-ray diffraction patterns of three Fe–17 at%Ga sintered samples and as-spun ribbon sample are shown in Figs. 4(a)–(c) and (d), respectively. The three sintered samples had the body-centered cubic structure with the lattice constant of  $0.2904$  nm. The intensity of the 200 peak of the samples sintered under 50 and 100 MPa at 973 K was similar to that of the as-spun ribbon sample which was manufactured from raw starting material. Figure 3(c) shows a scanning electron microscopic micrograph of the cross section of the sample sintered under 100 MPa at 973 K. A columnar grain size of approximately  $10\ \mu\text{m}$  parallel to the thickness direction was clearly observed. These results suggest that this sintered sample maintained the  $[001]$ -oriented texture of ribbon after sintering. The same tendency also existed in the sample sintered under 50 MPa at 973 K.

On the other hand, the 200 peak of the sample sintered

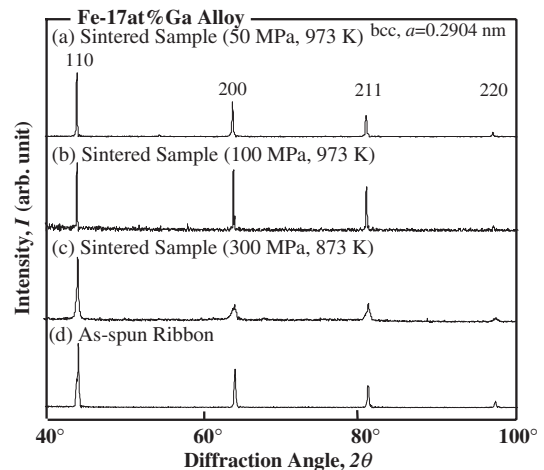


Fig. 4 X-ray diffraction patterns of Fe–17 at%Ga sintered samples and as-spun ribbon sample.

under 300 MPa at 873 K was weakened and broadened, indicating random orientation similar to the case of the bulk sample. The high pressure of 300 MPa caused plastic deformation and internal damage in the sample during sintering, which enhanced the driving energy for nucleation and subsequent recrystallization of grains. Accordingly, it was difficult for the 300 MPa sample to retain the texture of the as-spun ribbon.

The intensity ratios of the 200 to 110 peaks,  $I_{200}/I_{110}$ , for the sintered, bulk, and ribbon samples are listed in Table 1.

Next, it should be noted that the ordered phase such as  $L1_2$ , as shown in Fig. 2(a) in the case of general bulk material with the equilibrium phase of Fe–17 at%Ga alloy, did not appear after the SPSJ process. This fact illustrated that use of the SPSJ processing method to fabricate magnetostrictive bulk

Table 1 X-ray intensity ratio of Fe-17at%Ga alloys.

Sample	$I_{200}/I_{110}$
Bulk	0.21
As-spun Ribbon	0.57
Sintered Sample (50 MPa, 973 K)	0.55
Sintered Sample (100 MPa, 973 K)	0.80
(After Annealing)	(1.60)
Sintered Sample (300 MPa, 873 K)	0.14

Fe-17at%Ga alloy from laminates of rapidly solidified ribbons would work to suppress normal quasi-static diffusion due to its unique dynamic sintering mechanism that uses very large pulses of electrical current.

In consequence, these results showed that the [001] oriented texture and non-equilibrium phase of the as-spun ribbon were maintained in the samples sintered under the compressive stresses between 50 and 100 MPa using the SPSJ process.

### 3.1.2 The effect of heat treatment

The sample sintered under 100 MPa at 973 K was annealed for 1 h at 1173 K in order to release the internal stresses and sharpen the texture. The intensity of the 200 peak in the XRD pattern of the annealed sample became stronger, as listed in Table 1. This indicated that the [001] texture of this sintered sample was enhanced by annealing for a short time.

### 3.2 Magnetization and magnetostriction

The sintered Fe-17 at%Ga samples were cut to 2.7 mm in length, 5 mm in width and 9 mm in thickness, where the thickness is in the direction normal to the ribbon plate. When the magnetic field was applied in this direction of the ribbon, large magnetostriction resulted.<sup>6)</sup> In this study, the magnetic field  $H$  was applied in the thickness direction during the measurements of magnetization and magnetostriction.

Figures 5 and 6 show magnetization and magnetostriction respectively as a function of magnetic field for the sample sintered under 100 MPa at 973 K. As shown in Fig. 5, the

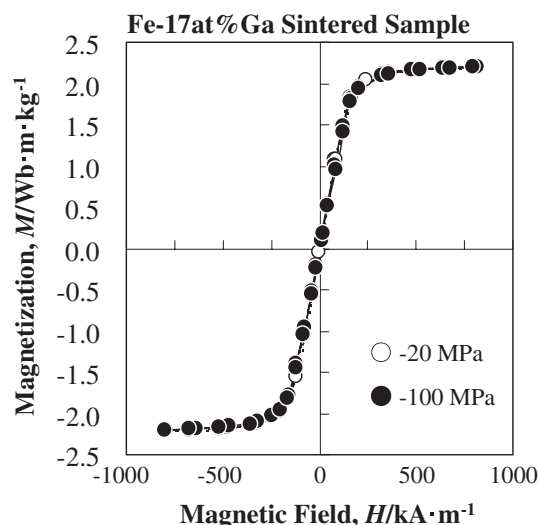


Fig. 5 Magnetization of Fe-17 at%Ga sintered sample (100 MPa, 973 K) with compressive pre-stresses of -20 and -100 MPa.

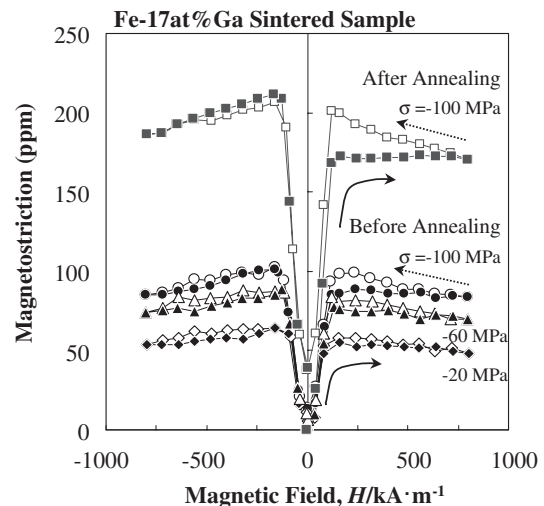


Fig. 6 Magnetostriction of Fe-17 at%Ga sintered sample (100 MPa, 973 K) before annealing with compressive pre-stresses of -20, -60 and -100 MPa, and after annealing at 1173 K for 1 h with pre-stress of -100 MPa.

magnetization was saturated at the applied magnetic field of 400 kA/m. The value of the saturation magnetization, 2.2 Wb·m/kg, was almost independent of compressive pre-stresses  $\sigma$ .

On the other hand, the magnetostriction was highly dependent on variations in  $\sigma$  and  $H$ . The magnetostriction for  $\sigma = -20$ ,  $-60$  and  $-100$  MPa increased rapidly and saturated at about  $H = |160|$  kA/m, and thereafter decreased slightly with further increase in  $H$ . When the magnitude of the magnetic field was reduced, the magnetostriction returned to its original value, generally along the initial path. The largest saturation magnetostriction of 100 ppm was obtained under  $\sigma = -100$  MPa.

The saturation magnetostriction data of the sintered samples are listed in Table 2. The sample sintered under 50 MPa at 973 K had the saturation magnetostriction of 70 ppm, which was smaller than that of the sample sintered under 100 MPa. This decrease for the sample sintered under 50 MPa could be attributed to the imperfect formation of bonding between the ribbons due to the lower stress applied during the sintering/joining process. On the other hand, the sample sintered under 300 MPa at 873 K, whose crystal orientation became random, also had small magnetostriction.

From these facts, in order to fabricate large magnetostrictive material, the lamination of ribbons must be joined tightly. Since the surface of the ribbon was uneven and rough, full and complete junction between the laminated ribbons may not have been achieved in the test samples. However, we consider that the sample sintered under 100 MPa at 973 K

Table 2 Saturation magnetostriction of sintered Fe-17 at%Ga samples.

SPS Condition	Magnetostriction
50 MPa, 973 K	70 ppm
100 MPa, 973 K	100 ppm
(After Annealing)	(170 ppm)
300 MPa, 873 K	60 ppm

was successful in achieving full and complete junction between the laminated ribbons, as it exhibited almost the same density as the Fe–17 at%Ga alloy ingot.

Next, the effect of heat treatment on magnetostriktion of the sample sintered under 100 MPa at 973 K was studied. The magnetostriktion of this laminated sample after annealing for 1 h at 1173 K is shown in Fig. 6. The magnetostriktion before and after annealing at 160 kA/m was 100 and 170 ppm, respectively. The magnetostriktion at the positive and negative magnetic field also gave the same value. However, the start point at the negative field shifted up, and the curve of magnetostriktion moved up due to the retained magnetostriktion. The effect of annealing for a short time made magnetostriktion increase, which resulted from the stronger [001]-oriented texture in the laminate ribbons as listed in Table 1.

From these results, it was confirmed that polycrystalline Fe–Ga bulk alloy with large magnetostriktion could be fabricated by combining the high performance rapid-solidified material element and the SPSJ processing method under the following conditions: (1) the laminated ribbons were sintered under 100 MPa at 973 K for 5 min, and (2) the sintered laminate was annealed for a short time.

#### 4. Conclusions

We have produced sintered bulk Fe–17 at%Ga laminates by applying the SPSJ fabrication method to rapid-solidified ribbons. The laminated and subsequently sintered sample joined by the SPSJ fabrication under pressure of 100 MPa at a temperature of 973 K retained the polycrystalline [001] oriented texture with the columnar grains of the as-spun ribbons. The ordered  $L1_2$  phase, which is generally formed in melt-worked samples, was scarcely observed in the as-spun

raw ribbon, as well as in the laminated bulk sample produced by the SPSJ method because of its unique dynamic and short-time sintering at rather low temperature. Magnetization was saturated at an applied magnetic field of 400 kA/m. Magnetostriktion of 170 ppm, which was the same value as produced by the ribbon, arose at compressive pre-stress of –100 MPa and a magnetic field of about 160 kA/m.

The process proposed in this study to develop bulk actuator/sensor materials with the capability of delivering a large actuator force by combining the rapid-solidification technique and subsequent SPSJ fabrication method should prove effective in the manufacture of a new type of iron-based polycrystalline magnetostriktive material for engineering applications.

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